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Health risk from fluoride exposure of a population in selected areas of Tamil Nadu South India

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Abstract

Prevalence of fluorosis is a public health problem in many states of India. It is necessary to find out the different sources through which fluoride enters human metabolism. Only when the sources are identified, suitable remedial measures can be initiated. This study was attempted to find out the contribution of fluoride from various sources such as drinking water, staple food grains, cooked rice, green leafy vegetables and cow milk in the selected area of the study. Percentage of incidence of fluorosis was estimated using a clinical survey. Calculated community fluorosis index values in all the 22 villages were greater than the accepted index value. Fluoride intake from all manor dietary sources of different age groups such as infants, children, adolescents, adults and aged above 70 was determined. The study revealed that, of all the sources, fluoride contribution from drinking water is significant irrespective of age. Hence, it is advised that people of South India, where there similarity in diet pattern, consume drinking water with lesser fluoride to minimize the debilitating effect of fluoride. The study also recommends to the government authorities concerned with supply to provide water with low fluoride level.

Keywords: Community fluorosis index; Total fluoride intake; Fluoride exposure level; Dietary sources

1. Introduction

About 80% of the diseases in the world are due to poor quality of drinking water, and the fluoride contamination in drinking water is responsible for 65% of endemic fluorosis in the world [1–3]. Fluorosis is a slow, progressive, crippling malady, which affects every organ, tissue and cell in the body and results in health complaints having overlapping manifestations with several diseases. The disease "fluorosis" has now become a global problem and the health impairment due to fluorosis has occurred in the citizens of about 25 nations across the globe, and more than 200 million people worldwide are at the risk of fluorosis [4]. Fluoride toxicity depends for its severity on four factors (i.e.) the total dose ingested, duration of fluoride exposure, nutritional status and body's response [5].

Different forms of fluorosis are dental, skeletal and non-skeletal. Dental fluorosis is a developmental disturbance of dental enamel caused by excessive exposure to high concentrations of fluoride during tooth development. The risk of fluoride overexposure occurs between the ages of 3 months and 8 years. In its mild forms fluorosis often appears as unnoticeable, tiny white streaks or specks in the enamel of the tooth. In its most severe form, tooth appearance is marred by discoloration or brown markings. The enamel may be pitted, rough and hard to clean. The spots and stains left by fluorosis are permanent and may darken over time. The severity of dental fluorosis depends on the amount of fluoride exposure, the age of the child, individual response, weight, degree of physical activity, nutrition, and bone growth. Skeletal fluorosis is a bone disease caused by excessive consumption of fluoride. It causes pain and damage to bones and joints. Fluoride when consumed in excess, can also affect non-calcified tissues besides bone and teeth is called as non skeletal fluorosis. The soft tissue organs affected by fluoride are named in the following order: aorta, thyroid, lungs, kidneys, heart, pancreas, brain and spleen [6].

Fluoride is readily absorbed from the gastro-intestinal tract, with estimates of absorption ranging from 75% to 100% [7,8]. The systemic fluoride absorption from water through the gastrointestinal tract into blood stream is nearly 100% by the process

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of simple diffusion without any intervention of overall water quality [9–11]. Fifty percent of the ground water sources in India have been contaminated by fluoride and more than 90% of rural drinking water supply programs are based on ground water [12]. Fluoride intake from water depends on the amount of water ingested through itself and the quantity of water ingested through food by means of water used for cooking and their fluoride content [13]. Many previous studies, from various parts of the world reported the development of dental fluorosis even if the people consume drinking water with fluoride less than 1.0 mg/L [14,15], which implies that the optimal fluoride dose level in drinking water may vary with various features such as local climatic conditions, methods of food processing and cooking [16], amount of food and water intake and its fluoride and other nutrients level, and dietary habits of the community [17–19].

In India, there are only limited studies available in the literature on fluoride content of raw foods and it became clear that fluorosis varies within the population. Factors responsible for these variations could be fluoride intake by drinking water, dietary intake, especially intake of food grown in soil or irrigated with water rich in fluoride. Therefore, the present study attempts to find out the fluoride content of drinking water, staple food grains, green leafy vegetables, cow milk samples and also cooked rice predominantly consumed by the people of certain fluoride endemic areas of Tamil Nadu, South India and to correlate estimated daily intake with dental fluorosis. The exposure doses of fluoride from the consumption of vegetable and cereal crops in children (3–14 years of age) are found to be higher than the limits laid down by Institute of Medicine, Washington, DC [20]. This means the children in this age group are more likely at the risk of fluorosis from consumption of vegetables and cereal grown in the fluoridated areas.

The aim of the study is to find out the average fluoride intake from various sources and to correlate with the percentage prevalence of fluorosis. Identification of the source which contributes more is to be specified so that effective interventions can be suggested to the people of the affected areas. In order to find out the extent of fluoride contamination in drinking water and to find out the fluoride exposure dose in Nilakottai block, an extensive study was accomplished by estimating fluoride level in drinking water.

2. Materials and methods

2.1. Selection of the study areas

The criteria for selection of the study area are the high percentage prevalence of dental fluorosis and the presence of fluoride in excess of 1 mg/L in drinking water samples. The percentage of dental fluorosis was estimated based on a clinical survey carried out among school children above 7 years (the age above which milk teeth are replaced by permanent ones) and among village adults. The villages were selected from the Nilakottai block of Dindigul district. Goggle earth satellite image of exact locations of fluoride endemic villages were prepared and is shown in Fig. 1.

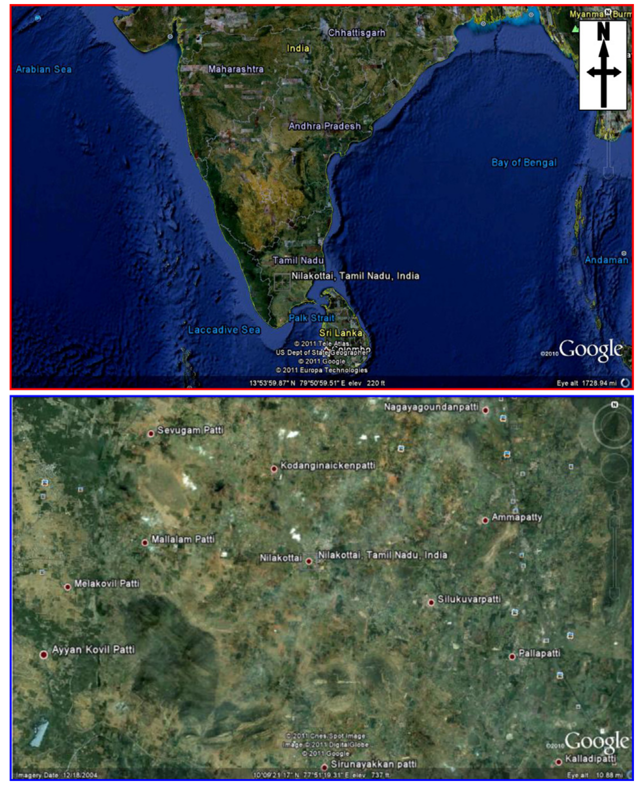

Fig. 1. Location of fluoride endemic villages in Nilakottai block in Dindigul district.

2.2. Collection and analysis of drinking water samples

The study was carried out in 22 villages of the block 110 drinking water samples were collected from 22 villages in Nilakottai block, Dindigul district, Tamil Nadu, South India for fluoride analysis. Five drinking water samples were collected from each village of the selected areas and stored in clean, high density polyethylene bottles at 4 °C before being analyzed. Fluoride levels in drinking water samples were measured by using fluoride ion selective electrode Orion ion analyzer (Thermo scientific Orion 4 Star pH. ISE Benchtop) by diluting with total ionic strength adjustment buffer (TISAB III) in 10:1 ratio. The instrument was calibrated with standard fluoride solutions so chosen the concentration of one was ten times the concentration of the other and also that the concentration of the unknown falls between those standards. Then the concentration of the unknown was directly read from the digital display of the meter [21].

2.3. Collection and analysis of staple food grains

The chief staple food grains grown in the selected fluorotic villages were identified. A total of 528 grain samples cultivated in the selected fluorotic villages were collected. Eight different grains, normally consumed by the people were collected in triplicate of different fields of the selected areas using random sampling technique. To determine total fluoride in the grain samples, 20 g of dried and ground grain sample was taken in nickel crucible, to which, 10 mL calcium hydroxide suspension was added as a fluoride fixing agent and mixed thoroughly. The sample was evaporated to dryness in a hot air oven at 80 °C and then ashed in muffle furnace at 600 °C for 12 h. The ash was transferred to a distillation flask. Sufficient analar silver sulphate was added to precipitate any chloride present in the sample. Pieces of glass and porcelain were also put in the flask. 25 mL of 60% analar perchloric acid and 20 mL of distilled water were added and then distillation was carried out. The distillate collected between 135 °C and 139 °C, was neutralized using 0.2 mol/L sodium hydroxide and analyzed for fluoride using fluoride ion selective electrode [22].

2.4. Collection and analysis of solid food samples

A total of 66 cooked rice samples, three from each selected area were collected and analyzed as per the procedure adopted for food grain samples [22].

2.5. Collection and analysis of green leafy vegetables (GLV)

A total of 660 GLV samples grown and usually present in the diet of the people of selected area were collected, dried, ground and analyzed as per the standard procedure adopted [22]. Ten different varieties were collected depending on the availability in an area.

2.6. Collection and analysis of cow milk samples

Among a total of 66 cow milk samples, 3 from each selected area were collected in polyethylene bottles and refrigerated until the time of analysis. To determine fluoride concentration, 50 mL of the cow milk sample was taken in a distillation flask; sufficient silver sulphate was added to precipitate the chlorides present. 50 mL of analar 60% perchloric acid was added and the distillate was collected between 135 °C and 139 °C and it was neutralized by using 0.2 mol/L sodium hydroxide and analyzed for fluoride using fluoride ion selective electrode [22].

2.7. Clinical survey and community fluorosis index (CFI)

Clinical survey was conducted among the people living in the selected areas. Dental examinations were made using dental mirrors and probes under indirect sunlight by two qualified and well trained dentists. Community fluorosis index was calculated based on the symptoms of dental fluorosis using randomized sampling method, which is classified into seven categories based on Dean's classification viz., normal, questionable, very mild, mild, moderate, moderately severe and severe and each of these seven classifications were given a numerical weight such as 0.0, 0.5, 1.0, 1.5, 2.0, 3.0 and 4.0, respectively [23,24].

Criteria for Dean's fluorosis index are described as follows: Normal (0.0): the enamel represents the usual translucent semivitriform type of structure, and the surface is smooth, glossy, and usually of a pale creamy white color; Questionable (0.5): the enamel discloses slight aberrations from the translucency of normal enamel, ranging from a few white flecks to occasional white spots. This classification is utilized in those instances where a definite diagnosis of the mildest form of fluorosis is not warranted and a classification of "normal" is not justified; Very mild (1.0): small opaque, paper white areas scattered irregularly over the tooth but not involving as much as 25% of the tooth surface. Frequently included in this classification are teeth showing no more than about 1–2 mm of white opacity at the tip of the summit of the cusps of the bicuspids or second molars; Mild (1.5): the white opaque areas in the enamel of the teeth are more extensive but do not involve as much as 50% of the tooth; Moderate (2.0): all enamel surfaces of the teeth are affected, and the surfaces subject to attrition show wear. Brown stain is frequently a disfiguring feature; Severe: includes teeth formerly classified as "moderately severe (3.0)" and "severe (4.0)." All enamel surfaces are affected and hypoplasia is so marked that the general form of the tooth may be affected. The major diagnostic sign of this classification is discrete or confluent pitting. Brown stains are widespread and the teeth often present a corroded-like appearance [23–25].

People with symptoms of dental fluorosis were identified and classified in each category and the number of people in each category is multiplied by the corresponding numerical weight given in the brackets, the products thus obtained for the various categories are added up and the sum total divided by the total number of people surveyed, gives community fluorosis index. Only when the community fluorosis index value is greater than 0.6, fluorosis is considered to be a public health problem in that area [23,26].

$$\mathrm{CFI} = \sum \frac{\text{Number of people} \times \text{Dean's numerical weight}}{\text{Total number of people}}$$

The percentage incidence of fluorosis was estimated from the number of people affected by fluorosis from the particular area with total number of people surveyed.

2.8. Nutritional survey

The nutritional survey among the population of different age groups infants, children and adults was conducted in the selected endemic fluoride area. Based on household survey, quantity of drinking water, cow milk, GLVs, food grains and cooked food consumed per day by the various age groups of people and their body weight were accounted in the selected villages and the values were verified with the data available with the local primary health centers.

2.9. Estimation of total fluoride intake (TFI)

Total fluoride intake (TFI) is the summation of the daily fluoride intake through entire diet sources. Daily fluoride intake (DFI) from the particular diet source was calculated by multiplying the fluoride concentration of the respective item with total quantity of the particular item consumed per day.

$$\mathrm{TFI} = \sum(\mathrm{DFI})$$

$$DFI = FC \times QD$$

where FC is the fluoride concentration in the diet source and QD is the quantity of the diet intake per day.

2.10. Fluoride exposure dose through drinking water (ED)

The fluoride exposure dose through drinking water was calculated by the following equation:

$$\text{Fluoride exposure dose} = \frac{FC \times WI}{BW}$$

where FC is the fluoride concentration (mg/L), WI is the water intake (L/day) and BW is the body weight (kg).

The assumption behind the calculation is persistent exposure and total bioavailability of fluoride concentration in water. The water intake of different age groups was estimated from the nutritional survey. Infants in their emergent life drink 250 mL of water as boiled water per day. In boiled water, fluoride level increases proportionally to the loss of volume, so the fluoride concentration in the actual drinking water was doubled [16]. The estimated water intake for the infant, children and adult was 0.25, 1.50 and 3.0 L per day, respectively. For the calculation, body weight of infants in the age group of 0–6 years was kept as 6 kg and children between 7 years and 19 years as 25 kg body weight and that of adults above 19 years as 60 kg body weight, the mean of water fluoride level in each village was used for fluoride exposure dose calculation.

2.11. Statistical analysis

Fluoride concentrations in water and diet sources such as grains, cooked food, green leafy vegetable and cow milk were expressed as mean and standard deviation of samples and the values are presented. The extend of linearity and correlation between water fluoride level with contribution of various diet sources was estimated using correlation coefficient (r) and coefficient of determination (R^2). The quantity of numerical relationships between the total fluoride intake and the contribution of diet sources were expressed through regression analysis and were performed by using SPSS Inc, Origin Pro and Excel.

3. Results

3.1. Fluoride concentration in drinking water and CFI of the study area

All samples from 22 selected fluoride endemic villages contain more than 1.5 mg/L fluoride (Table 1). Among these villages, Akkarakaranpatti, Silukuvarpatti and Thoppinayakkanpatti contain more than 3 mg/L fluoride, which is three times higher than the safe fluoride level in drinking water. 14% of drinking water samples of the selected villages have more than 3 mg/L of fluoride, remaining 86% drinking water samples have more than 1.5 mg/L fluoride. Higher prevalence rate of fluorosis was observed from Akkarakaranpatti and Thoppinayakkanpatti that are 82.4% and 81.8%, respectively, with 1.57 and 1.34 CFI values. The numerical relationship between the water fluoride level with percentage of fluorosis prevalence and community fluorosis indices of the study areas indicated that, CFI values of all the selected endemic villages are higher than 0.6, a value above which fluorosis is considered to be a public health problem. Community fluorosis index and the percentage of fluorosis prevalence from all the selected endemic villages were shown in Table 2. The percentage of prevalence of

Table 1
Drinking water fluoride levels in selected fluoride endemic areas of Nilakottai block.

No.	Name of the village	Levels of fluoride (mg/L)					Mean ± SD	Range
		East	West	North	South	Middle		
1	Alagampatti	2.0	1.8	1.8	1.9	2.1	1.92 ± 0.13	1.8–2.1
2	Sangalpatti	1.5	1.7	1.5	1.5	1.5	1.54 ± 0.09	1.5–1.7
3	Micheilpalayam	2.1	2.3	2.1	2.2	2.2	2.18 ± 0.08	2.1–2.3
4	Ammapatti	1.8	2.0	1.8	1.9	1.9	1.88 ± 0.08	1.8–2.0
5	Uchanampatti	1.8	1.7	1.6	1.9	1.7	1.74 ± 0.11	1.6–1.9
6	Othur	1.7	1.5	1.6	1.5	1.7	1.60 ± 0.10	1.5–1.7
7	Chockanchettipatti	1.9	2.2	1.8	1.9	1.8	1.92 ± 0.16	1.8–2.2
8	Murugathuranpatti	1.5	1.8	1.5	1.8	1.6	1.64 ± 0.15	1.5–1.8
9	Kulalakundu	1.8	1.6	1.9	1.5	1.5	1.66 ± 0.18	1.5–1.9
10	Sandilarpuram	1.6	1.7	1.6	1.8	1.6	1.66 ± 0.09	1.6–1.8
11	Nagayakavundampatti	1.9	1.8	2.0	2.2	1.8	1.94 ± 0.17	1.8–2.2
12	Kolinjipatti	2.2	2.4	2.0	1.9	2.1	2.12 ± 0.19	1.9–2.4
13	Thoppinayakanpatti	3.4	2.9	3.1	3.1	3.2	3.14 ± 0.18	2.9–3.4
14	Meenachipuram	1.7	2.0	1.7	1.8	1.9	1.82 ± 0.13	1.7–2.0
15	Pallapatti	1.8	1.6	2.0	1.6	1.7	1.74 ± 0.17	1.6–2.0
16	Kattukothampatti	1.6	1.5	1.5	1.6	1.4	1.52 ± 0.08	1.4–1.6
17	Kanthappankottai	1.5	1.7	1.4	1.4	1.7	1.54 ± 0.15	1.4–1.7
18	Sangarapuram	2.7	2.4	2.1	2.2	2.4	2.36 ± 0.23	2.1–2.7
19	Silukuvarpatti	3.6	3.1	3.4	3.2	3.2	3.30 ± 0.20	3.1–3.6
20	Akkarakaranpatti	3.4	3.2	3.5	3.1	3.4	3.32 ± 0.16	3.1–3.5
21	Sithargalnatham	1.9	1.8	2.0	1.8	2.1	1.92 ± 0.13	1.8–2.1
22	Kullichettipatti	1.8	1.6	1.6	1.7	1.8	1.70 ± 0.10	1.6–1.8

Table 2
Community fluorosis index and percentage prevalence of fluorosis in the area of study.

Name of the village	Water fluoride level (mg/L), mean ± SD	Normal	Questionable	Very mild	Mild	Moderate	Moderate severe	Severe	Total	CFI	% of DF
		0	0.5	1	1.5	2	3	4			
Alagampatti	1.92 ± 0.13	32	16	18	14	30	16	8	134	1.19	67.80
Sangalpatti	1.54 ± 0.09	38	19	13	16	21	14	3	124	0.97	64.49
Micheilpalayam	2.18 ± 0.08	33	15	11	18	24	16	6	123	1.41	72.73
Ammapatti	1.88 ± 0.08	41	12	13	16	14	12	2	110	1.30	66.94
Uchanampatti	1.74 ± 0.11	38	22	16	12	16	11	2	117	0.97	66.37
Othur	1.60 ± 0.10	35	15	13	17	6	4	2	92	0.98	63.92
Chockanchettipatti	1.92 ± 0.16	47	21	23	12	18	10	3	134	1.03	67.13
Murugathuranpatti	1.64 ± 0.15	43	26	15	11	21	14	4	134	0.93	64.46
Kulalakundu	1.66 ± 0.18	32	17	21	14	22	18	3	127	1.00	64.75
Sandilarpuram	1.66 ± 0.09	34	14	11	14	18	13	2	106	1.11	65.45
Nagayakavundampatti	1.94 ± 0.17	43	28	18	15	12	13	1	130	1.07	67.81
Kolinjipatti	2.12 ± 0.19	32	22	11	16	18	26	6	131	1.31	72.18
Thoppinayakanpatti	3.14 ± 0.18	21	18	12	12	15	11	3	92	1.34	81.82
Meenachipuram	1.82 ± 0.13	45	14	21	16	22	11	4	133	1.10	66.92
Pallapatti	1.74 ± 0.17	38	21	26	22	33	21	6	167	1.08	66.67
Kattukothampatti	1.52 ± 0.08	36	12	10	17	13	11	2	101	1.05	63.27
Kanthappankottai	1.54 ± 0.15	28	10	8	11	13	5	1	76	1.02	63.64
Sangarapuram	2.36 ± 0.23	27	22	18	25	21	18	8	139	1.27	75.69
Silukuvarpatti	3.30 ± 0.20	38	28	32	31	37	24	11	201	1.19	75.40
Akkarakaranpatti	3.32 ± 0.16	23	22	18	15	17	22	10	127	1.57	82.44
Sithargalnatham	1.92 ± 0.13	28	20	12	10	12	10	1	93	1.07	67.31
Kullichettipatti	1.70 ± 0.10	32	10	11	14	10	8	1	86	1.09	64.04

NOTE: % of DF: percentage of dental fluorosis.

Table 3
Correlation between total fluoride intake and related variables.

Variables	r	R^2	Regression equation	95% confidence interval for r	Significant level (P)	F ratio
% incidence of fluorosis	0.9770	0.9545	$45.9776 + 11.5782x$	0.9444–0.9906	<0.0001	419.49
Community fluorosis index	0.8404	0.7062	$0.5656 + 0.2927x$	0.6485–0.9318	<0.0001	48.07
Water fluoride level	0.9991	0.9982	$0.2042 + 0.3264x$	0.9914–0.9999	<0.0001	2155.58

fluorosis and community fluorosis index increased significantly ($r = 0.977$, $R^2 = 0.955$, $P < 0.0001$) with the increase in drinking water fluoride level. The quantitative numerical relationship between the water fluoride level with percentage of fluorosis prevalence and community fluoride indices is shown in Table 3. Almost 30% of the people in the selected fluoride endemic villages are affected with more than mild form of fluorosis with higher CFI values.

3.2. Correlation analysis between fluoride concentration in drinking water and maximum exposure dose

Correlation between the water fluoride levels with exposure dose of different age groups is shown in Table 4. High degree of correlation is obtained ($r = 0.99$, $R^2 = 0.99$) for all age groups. The significant levels $P < 0.0001$ and the numerical relationship and the regression equations between the drinking water fluoride level and fluoride exposure dose are listed in Table 4. The obtained R^2 and analysis of variance indicate a high significant relationship between drinking water fluoride levels with the maximum exposure dose. The correlation coefficient and regression equations give the high significant linearity and numerical relationship between water fluoride levels with exposure dose.

3.3. Fluoride concentration in milk samples, green leafy vegetables, food grains and cooked foods

The concentrations of fluoride in samples of milk, green leafy vegetables, food grains and cooked foods collected from selected endemic areas with varying fluoride concentration in ground

Table 4
Correlation between water fluoride level and exposure dose.

Variables	r	R^2	Regression equation	95% confidence interval for r	Significant level (P)	F ratio
Infants	0.9980	0.9960	$0.0019 + 0.0829x$	0.9951–0.9992	<0.0001	5020.51
Children	0.9952	0.9904	$-0.0018 + 0.0612x$	0.9882–0.9980	<0.0001	2064.89
Adults	0.9941	0.9882	$-0.0013 + 0.0516x$	0.9855–0.9976	<0.0001	1672.59

Table 5
Fluoride levels of cow milk samples in selected fluoride endemic areas.

Name of the village	Levels of fluoride (mg/L), mean ± SD
Alagampatti	0.048 ± 0.005
Sangalpatti	0.044 ± 0.003
Micheilpalayam	0.058 ± 0.006
Ammapatti	0.050 ± 0.005
Uchanampatti	0.053 ± 0.006
Othur	0.066 ± 0.004
Chockanchettipatti	0.053 ± 0.006
Murugathuranpatti	0.046 ± 0.011
Kulalakundu	0.045 ± 0.003
Sandilarpuram	0.050 ± 0.004
Nagayakavundampatti	0.059 ± 0.011
Kolinjipatti	0.057 ± 0.006
Thoppinayakanpatti	0.147 ± 0.006
Meenachipuram	0.067 ± 0.006
Pallapatti	0.057 ± 0.005
Kattukothampatti	0.048 ± 0.006
Kanthappankottai	0.043 ± 0.005
Sangarapuram	0.117 ± 0.006
Silukuvarpatti	0.120 ± 0.010
Akkarakaranpatti	0.113 ± 0.012
Sithargalnatham	0.077 ± 0.005
Kullichettipatti	0.059 ± 0.009

water are given in Tables 5–8. The results show that all the cow milk samples analyzed found to have very low levels of fluoride. Fluoride levels in cow milk ranged from 0.043 mg/L to 0.147 mg/L in the selected fluoride endemic villages. All the varieties of green leafy vegetables grown in Sangarapuram, Thoppinayakanpatti, Silukuvarpatti and Akkarakaranpatti found to have high content of fluoride. An interesting observation is that, in all the areas, the *Acalypha indica* leaves recorded high fluoride level, but the majority of the people living in these endemic fluoride areas use *Moringa oleifera* and *Sesbania grandiflora* in their regular diet as per the nutritional survey data. Both *Moringa oleifera* and *Sesbania grandiflora* also contain higher fluoride concentrations as shown in Table 6. In Akkarakaranpatti village the fluoride concentration in *Moringa oleifera* and *Sesbania grandiflora* were recorded as 7.68 mg/kg and 4.47 mg/kg, respectively, and in the Thoppinayakanpatti village the fluoride concentration in *Moringa oleifera* and *Sesbania grandiflora* were recorded as 6.77 mg/kg and 4.88 mg/kg, respectively. Fluoride concentrations of food grain samples were analyzed and the results are given in Table 7. It was found that green gram, finger millet, pearl millet, field been and black-eyed bean contained the maximum amount of fluoride. From the household survey, it was observed that, most of the people use black-eyed bean and field bean in their regular diet. Both the food grains also contain more fluoride concentration in all selected villages. Fluoride content in cooked food samples collected from selected villages was analyzed and it is given in Table 8. Samples from Akkarakaranpatti, Sankarapuram and Silukuvarpatti registered high content of fluoride. Food grains particularly black-eyed bean recorded 4.21, 4.12, 4.12, 4.11 mg/kg of fluoride content in Silukuvarpatti, Sankarapuram, Pallapatti and Akkarakaranpatti, respectively, and the field bean samples from

Table 6
Fluoride levels of green leafy vegetables in selected fluoride endemic areas.

Name of the village	Levels of fluoride (mg/kg), mean ± SD									
	Sesbania grandiflora	*Moringa oleifera*	*Solanum nigrum*	*Amaranthus spinosus*	*Amaranthus polygonoides*	*Rumex acetosa*	*Alternanthera sessilis*	*Amaranthus dubius*	*Acalypha indica*	*Basella alba*
Alagampatti	2.79 ± 0.15	2.32 ± 0.14	1.63 ± 0.34	1.97 ± 0.13	2.24 ± 0.11	1.53 ± 0.29	1.75 ± 0.08	1.38 ± 0.15	3.86 ± 0.10	1.62 ± 0.09
Sangalpatti	2.45 ± 0.20	1.45 ± 0.10	1.45 ± 0.23	1.37 ± 0.10	1.84 ± 0.08	1.55 ± 0.25	1.87 ± 0.25	1.17 ± 0.10	3.48 ± 0.29	1.65 ± 0.13
Micheilpalayam	3.49 ± 0.24	2.75 ± 0.12	1.58 ± 0.22	1.74 ± 0.13	1.60 ± 0.20	1.75 ± 0.09	1.84 ± 0.09	1.52 ± 0.30	4.16 ± 0.11	1.80 ± 0.10
Ammapatti	2.59 ± 0.29	2.32 ± 0.06	1.87 ± 0.08	2.04 ± 0.11	2.37 ± 0.13	1.46 ± 0.07	1.90 ± 0.16	1.56 ± 0.07	4.16 ± 0.11	1.64 ± 0.09
Uchanampatti	2.60 ± 0.20	2.20 ± 0.16	1.24 ± 0.05	1.67 ± 0.14	2.05 ± 0.13	1.30 ± 0.07	1.63 ± 0.10	1.48 ± 0.20	3.23 ± 0.19	1.40 ± 0.11
Othur	2.47 ± 0.08	3.68 ± 0.23	1.10 ± 0.16	0.74 ± 0.09	1.02 ± 0.06	1.02 ± 0.07	0.96 ± 0.12	0.88 ± 0.04	0.58 ± 0.04	0.62 ± 0.04
Chockanchettipatti	2.48 ± 0.27	1.53 ± 0.25	1.24 ± 0.14	1.54 ± 0.27	1.68 ± 0.06	1.70 ± 0.57	1.92 ± 0.13	1.33 ± 0.21	2.58 ± 0.04	1.13 ± 0.12
Murugathuranpatti	2.06 ± 0.12	1.74 ± 0.10	1.25 ± 0.03	1.63 ± 0.21	1.58 ± 0.04	1.36 ± 0.08	1.70 ± 0.15	1.09 ± 0.07	2.37 ± 0.24	1.22 ± 0.12
Kulalakundu	2.32 ± 0.17	2.23 ± 0.06	2.04 ± 0.22	2.35 ± 0.13	2.58 ± 0.04	1.20 ± 0.16	2.15 ± 0.09	1.79 ± 0.09	3.60 ± 0.21	1.66 ± 0.20
Sandilarpuram	2.68 ± 0.15	1.97 ± 0.13	1.26 ± 0.04	1.65 ± 0.15	2.27 ± 0.12	1.46 ± 0.20	1.51 ± 0.12	1.41 ± 0.12	3.48 ± 0.13	2.26 ± 0.08
Nagayakavundampatti	2.45 ± 0.24	2.12 ± 0.09	1.65 ± 0.09	1.66 ± 0.12	2.39 ± 0.10	1.63 ± 0.13	2.07 ± 0.10	1.62 ± 0.09	3.31 ± 0.16	1.76 ± 0.09
Kolinjipatti	3.26 ± 0.17	2.58 ± 0.23	1.60 ± 0.29	2.05 ± 0.09	2.17 ± 0.12	2.16 ± 0.09	1.98 ± 0.12	1.72 ± 0.04	3.86 ± 0.11	2.30 ± 0.15
Thoppinayakanpatti	4.88 ± 0.34	6.77 ± 0.51	5.09 ± 0.36	6.13 ± 0.37	5.69 ± 0.51	3.56 ± 0.32	4.41 ± 0.23	5.00 ± 0.17	5.18 ± 0.08	3.51 ± 0.22
Meenachipuram	2.41 ± 0.19	2.30 ± 0.12	1.30 ± 0.05	1.49 ± 0.19	1.17 ± 0.08	1.37 ± 0.17	1.91 ± 0.16	1.47 ± 0.05	3.40 ± 0.06	1.66 ± 0.14
Pallapatti	2.67 ± 0.20	2.26 ± 0.10	1.61 ± 0.20	2.59 ± 0.33	2.51 ± 0.14	2.04 ± 0 .11	2.02 ± 0.10	1.51 ± 0.09	3.43 ± 0.14	2.06 ± 0.12
Kattukothampatti	2.38 ± 0.33	1.58 ± 0.30	0.92 ± 0.12	1.58 ± 0.25	2.35 ± 0.16	1.58 ± 0.10	1.96 ± 0.17	1.17 ± 0.09	1.30 ± 0.11	2.23 ± 0.12
Kanthappankottai	2.54 ± 0.13	1.53 ± 0.15	1.62 ± 0.28	1.68 ± 0.10	2.31 ± 0.17	1.61 ± 0.16	1.68 ± 0.23	1.14 ± 0.07	1.48 ± 0.03	1.60 ± 0.21
Sangarapuram	5.09 ± 0.19	5.48 ± 0.24	5.92 ± 0.09	5.56 ± 0.28	5.83 ± 0.25	4.43 ± 0.38	5.29 ± 0.17	4.75 ± 0.13	5.93 ± 0.17	4.32 ± 0.28
Silukuvarpatti	4.28 ± 0.09	5.57 ± 0.28	5.76 ± 0.20	4.76 ± 0.17	4.56 ± 0.29	4.29 ± 0.29	5.63 ± 0.17	5.41 ± 0.22	6.95 ± 0.23	6.68 ± 0.19
Akkarakaranpatti	4.47 ± 0.26	7.68 ± 0.32	5.48 ± 0.16	5.41 ± 0.20	6.39 ± 0.16	4.52 ± 0.21	5.79 ± 0.16	5.43 ± 0.19	4.79 ± 0.40	3.39 ± 0.30
Sithargalnatham	2.75 ± 0.12	2.17 ± 0.09	1.97 ± 0.21	1.76 ± 0.16	1.64 ± 0.19	1.77 ± 0.13	1.30 ± 0.07	1.19 ± 0.10	1.61 ± 0.19	1.59 ± 0.17
Kullichettipatti	3.29 ± 0.17	3.72 ± 0.13	2.03 ± 0.12	1.88 ± 0.21	1.96 ± 0.11	2.06 ± 0.09	1.56 ± 0.14	1.49 ± 0.05	1.48 ± 0.12	1.67 ± 0.15

Table 7
Fluoride levels of food grains in selected fluoride endemic areas.

Name of the village	Levels of fluoride (mg/kg), mean ± SD							
	Black-eyed bean	Field bean	Great millet	Finger millet	Pearl millet	Green gram	Horse gram	Maize
Alagampatti	3.14 ± 0.09	2.61 ± 0.22	2.30 ± 0.14	4.29 ± 0.08	3.22 ± 0.14	4.94 ± 0.12	2.61 ± 0.15	2.47 ± 0.17
Sangalpatti	1.57 ± 0.13	2.67 ± 0.19	2.15 ± 0.11	3.05 ± 0.17	2.62 ± 0.15	3.59 ± 0.12	2.48 ± 0.12	2.56 ± 0.19
Micheilpalayam	3.67 ± 0.13	2.95 ± 0.12	2.54 ± 0.18	3.86 ± 0.11	3.06 ± 0.12	3.52 ± 0.18	2.63 ± 0.12	3.30 ± 0.20
Ammapatti	2.76 ± 0.36	2.45 ± 0.19	2.07 ± 0.14	2.44 ± 0.20	3.43 ± 0.19	2.75 ± 0.14	2.49 ± 0.18	1.96 ± 0.15
Uchanampatti	2.69 ± 0.21	1.62 ± 0.12	2.03 ± 0.15	1.99 ± 0.63	2.35 ± 0.24	2.66 ± 0.17	2.31 ± 0.11	1.85 ± 0.09
Othur	3.95 ± 0.15	2.50 ± 0.24	2.52 ± 0.27	3.60 ± 0.16	2.87 ± 0.17	4.75 ± 0.07	3.44 ± 0.28	3.05 ± 0.20
Chockanchettipatti	2.52 ± 0.13	2.19 ± 0.21	2.48 ± 0.22	1.85 ± 0.08	2.67 ± 0.13	1.97 ± 0.17	2.27 ± 0.09	2.32 ± 0.15
Murugathuranpatti	2.19 ± 0.18	2.50 ± 0.13	2.06 ± 0.08	2.29 ± 0.16	3.59 ± 0.18	2.35 ± 0.24	2.07 ± 0.12	1.64 ± 0.18
Kulalakundu	1.90 ± 0.27	1.69 ± 0.04	1.59 ± 0.24	3.16 ± 0.10	2.76 ± 0.15	2.35 ± 0.11	2.27 ± 0.25	2.05 ± 0.15
Sandilarpuram	2.05 ± 0.17	2.41 ± 0.13	1.82 ± 0.10	2.17 ± 0.09	2.63 ± 0.18	3.29 ± 0.14	1.68 ± 0.13	1.72 ± 0.14
Nagayakavundampatti	1.89 ± 0.16	1.98 ± 0.15	1.55 ± 0.10	1.42 ± 0.20	1.62 ± 0.22	1.96 ± 0.30	2.22 ± 0.12	1.40 ± 0.12
Kolinjipatti	3.61 ± 0.15	2.56 ± 0.18	2.12 ± 0.06	3.41 ± 0.22	2.59 ± 0.28	3.31 ± 0.19	2.55 ± 0.11	2.61 ± 0.17
Thoppinayakanpatti	3.73 ± 0.08	3.11 ± 0.16	2.42 ± 0.12	3.52 ± 0.20	2.80 ± 0.17	4.11 ± 0.15	2.46 ± 0.09	3.18 ± 0.25
Meenachipuram	2.59 ± 0.28	1.66 ± 0.07	1.96 ± 0.07	2.65 ± 0.17	1.76 ± 0.12	2.04 ± 0.24	1.97 ± 0.20	1.57 ± 0.23
Pallapatti	4.12 ± 0.12	2.12 ± 0.14	2.28 ± 0.26	2.88 ± 0.10	3.90 ± 0.14	2.07 ± 0.23	1.97 ± 0.28	2.49 ± 0.13
Kattukothampatti	1.44 ± 0.23	2.34 ± 0.08	2.16 ± 0.11	2.37 ± 0.05	2.58 ± 0.11	2.62 ± 0.13	1.94 ± 0.27	2.51 ± 0.27
Kanthappankottai	1.77 ± 0.18	1.84 ± 0.10	2.29 ± 0.10	2.01 ± 0.23	2.29 ± 0.30	2.34 ± 0.13	2.17 ± 0.15	1.46 ± 0.19
Sangarapuram	4.12 ± 0.27	2.40 ± 0.25	2.08 ± 0.17	2.56 ± 0.28	3.09 ± 0.17	2.58 ± 0.13	1.30 ± 0.16	1.63 ± 0.25
Silukuvarpatti	4.21 ± 0.33	2.43 ± 0.22	2.11 ± 0.25	2.68 ± 0.06	3.80 ± 0.46	2.14 ± 0.15	1.65 ± 0.14	2.24 ± 0.13
Akkarakaranpatti	4.11 ± 0.12	1.33 ± 0.17	2.46 ± 0.14	3.55 ± 0.25	3.96 ± 0.21	1.98 ± 0.13	1.41 ± 0.15	2.24 ± 0.14
Sithargalnatham	1.16 ± 0.18	2.02 ± 0.28	1.95 ± 0.23	1.66 ± 0.45	2.46 ± 0.26	1.94 ± 0.26	2.17 ± 0.15	1.99 ± 0.17
Kullichettipatti	1.49 ± 0.21	1.98 ± 0.08	1.29 ± 0.08	1.45 ± 0.20	1.51 ± 0.21	1.78 ± 0.12	2.14 ± 0.17	1.49 ± 0.12

Thoppinayakanpatti, Micheilpalayam and Sangalpatti recorded 3.11, 2.95, 2.67 mg/kg of fluoride, respectively.

3.4. Impact of fluoride ion concentration in drinking water on total fluoride intake

Daily average intake level of various dietary sources by different age groups of people from the study areas as collected through nutritional survey is shown in Table 9. To calculate the range of fluoride intake of the various age groups of people in the fluorotic area, six villages from the selected area were selected. These six villages were selected based on the level of fluoride in drinking water. Two each from low, moderate and high fluorotic areas were selected and the values are given in Table 10 and shown in Fig. 2. The total intake level of fluoride was compared with the average contribution from various dietary sources. The major fraction of the total fluoride intake was derived from water. For children, about 70% of the total fluoride intake was derived from drinking water. High degree of correlation was obtained ($r = 0.9991$, $R^2 = 0.9982$, $P < 0.0001$) between total fluoride intake and water fluoride level as shown in Table 11.

3.5. Impact of fluoride ion concentration in cow milk on total fluoride intake

All the cow milk samples analyzed found to have very low levels of fluoride. Among these, milk samples from Thoppinayakanpatti and Silukuvarpatti which contain more than 0.12 mg/L fluoride was recorded as high within the samples analyzed. Fluoride levels in cow milk ranged from 0.043 mg/L to 0.147 mg/L in the selected fluoride endemic villages. Cow milk showed significant contribution on total fluoride intake ($r = 0.967$, $R^2 = 0.935$, $P = 0.0016$) due to fluoride contamination as shown in Table 11.

Table 8
Fluoride levels of cooked rice in selected fluoride endemic areas.

Name of the village	Levels of fluoride (mg/kg), mean ± SD
Alagampatti	0.57 ± 0.05
Sangalpatti	0.48 ± 0.04
Micheilpalayam	0.59 ± 0.06
Ammapatti	0.49 ± 0.04
Uchanampatti	0.43 ± 0.04
Othur	0.52 ± 0.04
Chockanchettipatti	0.34 ± 0.05
Murugathuranpatti	0.44 ± 0.04
Kulalakundu	0.43 ± 0.04
Sandilarpuram	0.53 ± 0.10
Nagayakavundampatti	0.42 ± 0.07
Kolinjipatti	0.49 ± 0.06
Thoppinayakanpatti	0.58 ± 0.04
Meenachipuram	0.51 ± 0.07
Pallapatti	0.52 ± 0.03
Kattukothampatti	0.43 ± 0.08
Kanthappankottai	0.40 ± 0.08
Sangarapuram	0.65 ± 0.19
Silukuvarpatti	0.59 ± 0.07
Akkarakaranpatti	0.73 ± 0.06
Sithargalnatham	0.43 ± 0.05
Kullichettipatti	0.54 ± 0.02

Table 9
Average total intake level of various dietary sources as per nutritional survey.

Age group and weight range	Dietary source	Average daily intake level of dietary source
Infants		
Birth to 6 months (2–6 kg)	Drinking water	200 mL
	Cow milk	300 mL
Above 6–12 months (6–10 kg)	Drinking water	150 mL
	Cow milk	200 mL
Children		
From 1 to 3 years (11–18 kg)	Drinking water	500 mL
	Cow milk	250 mL
	Greens	100 g
	Grains	50 g
	Rice	200 g
From 3 to 10 years (18–25 kg)	Drinking water	750 mL
	Cow milk	400 mL
	Greens	150 g
	Grains	100 g
	Rice	500 g
From 10 to 18 years (25–45 kg)	Drinking water	2000 mL
	Cow milk	400 mL
	Greens	250 g
	Grains	150 g
	Rice	750 g
Adult		
From 18 to 70 years (45–70 kg)	Drinking water	2000 mL
	Cow milk	350 mL
	Greens	250 g
	Grains	200 g
	Rice	1000 g
Old		
Above 70 years (50–70 kg)	Drinking water	1500 mL
	Cow milk	300 mL
	Greens	200 g
	Grains	150 g
	Rice	1000 g

3.6. Impact of fluoride ion concentration in green leafy vegetables (GLVs) on total fluoride intake

The fluoride concentrations in GLVs were determined and presented in Table 6. It was observed that *Acalypha indica* leaves grown in selected endemic villages had the maximum concentration of fluoride. From the nutritional survey carried out in these areas, most of the people living in these villages reported that they predominantly consume *Moringa oleifera* and *Sesbania grandiflora*. Both these GLVs when analyzed found to have high content of fluoride which also contributed significantly to high fluoride exposure level of people. The linear relationship between fluoride content of GLVs grown in fluorotic villages ($r = 0.9876$, $P = 0.0002$) and the total fluoride intake was established as shown in Table 11. GLVs are one of the major sources of daily fluoride intake. Daily fluoride consumption of teenagers from GLVs in low, medium and high fluoride endemic villages was found to be 0.38, 0.58, 1.66 mg/day, respectively. Green leafy vegetables contribute 13% of the fluoride intake within the sources analyzed.

3.7. Impact of fluoride ion concentration in food grain on total fluoride intake

Fluoride levels of food grain samples collected from the selected endemic fluorotic areas are shown in Table 7. From the nutritional survey, it was learnt that, most of the people living in these villages prefer to have black-eyed bean and field bean in their regular diet. The obtained statistical correlations in Table 11 illustrate a high linear relationship ($r = 0.9536$, $R^2 = 0.9094$, $P = 0.0032$) between the total fluoride intake and intake through food grains. Among the dietary sources analyzed contribution of food grain is 10%.

3.8. Impact of fluoride ion concentration in cooked rice on total fluoride intake

Cooked rice is another major source of daily fluoride intake by children as well as adult people. 0.34, 0.38, 0.50 mg/d of fluoride was consumed at an average from cooked rice by adults in low, medium and high fluorotic areas, respectively. The values ranged from 1.16 mg/kg to 4.94 mg/kg for various food grains. There is no definite pattern in the concentration of fluoride with respect to the variety or place. However, all the samples from villages like Nagayakavundampatti and Kullichettipatti registered relatively less fluoride in all the grains and samples from Alagampatti, Thoppinayakkanpatti and Silukuvarpatti recorded little higher values. Table 11 illustrates a high linear relationship ($r = 0.9939$, $R^2 = 0.9878$, $P < 0.0001$) between the total fluoride intake and fluoride from cooked rice.

4. Discussion

In view of the environmental and socio-economic conditions of the Indian sub continent, the desirable limit of the fluoride is set at 0.60–1.20 mg/L and maximum permissible limit in absence of any other source is set at 1.5 mg/L for drinking water [27]. In this study about 98% of the daily fluoride intake by infants through their diet sources was derived from the local drinking water sources. Children residing in the fluorotic areas acquire nearly double the amount of fluoride than the optimal level. Because of powder-based infant formulae reconstituted with water from high fluoride endemic area containing more than 0.5 mg/L of fluoride may provide a daily fluoride intake of above the threshold limit of 0.10 mg/kg [28–30]. Drinking water seems to be the major contributor of fluoride among all the sources analyzed.

Fluoride ingestion through food is comparatively less than through water. However, it cannot be neglected in the endemic areas because it will increase the fluoride burden in addition to water. Fluoride not only enters through water, but also through many edible items. Fluoride of food items depends upon the fluoride contents of soil and water used for irrigation. Thus, fluoride in water contributes significantly to the total exposure of an individual to this element, but not the only source of exposure. In certain areas, the major contribution comes from diet sources as in the case of Vranjska Banja where agricultural products including potatoes, beans, tomato, cucumbers, water melons,

Table 10
Total fluoride intake per day from each dietary source in selected fluoride endemic areas.

Age group and weight range	Dietary source	Average daily intake level of dietary source	Average daily fluoride intake from the diet source (mg/d)					
			Kattukuthampatti	Sangalpatti	Meenachipuram	Ammapatti	Silukuvarpatti	Akkarakaranpatti
Infants								
Birth to 6 months (2–6 kg)	Drinking water	200 mL	0.304	0.308	0.364	0.376	0.640	0.680
	Cow milk	150 mL	0.007	0.007	0.010	0.008	0.018	0.017
Above 6–12 months (6–10 kg)	Drinking water	300 mL	0.456	0.462	0.546	0.564	0.960	1.020
	Cow milk	200 mL	0.010	0.009	0.013	0.010	0.024	0.023
Children								
From 1 to 3 years (11–18 kg)	Drinking water	500 mL	0.760	0.770	0.910	0.940	1.600	1.700
	Cow milk	250 mL	0.012	0.011	0.017	0.013	0.030	0.283
	Greens	100 g	0.158	0.145	0.230	0.232	0.557	0.768
	Grains	50 g	0.072	0.079	0.129	0.138	0.211	0.206
	Rice	200 g	0.090	0.096	0.102	0.108	0.138	0.146
From 3 to 10 years (18–25 kg)	Drinking water	750 mL	1.140	1.155	1.370	1.410	2.400	2.550
	Cow milk	400 mL	0.019	0.018	0.027	0.020	0.048	0.045
	Greens	150 g	0.237	0.218	0.345	0.348	0.836	1.152
	Grains	100 g	0.144	0.157	0.259	0.276	0.421	0.411
	Rice	500 g	0.215	0.240	0.255	0.270	0.345	0.365
From 10 to 18 years (25–45 kg)	Drinking water	2000 mL	3.040	3.08	3.640	3.760	6.400	6.800
	Cow milk	400 mL	0.019	0.018	0.027	0.020	0.048	0.045
	Greens	250 g	0.395	0.363	0.575	0.580	1.393	1.920
	Grains	150 g	0.216	0.236	0.389	0.414	0.632	0.617
	Rice	750 g	0.338	0.360	0.383	0.405	0.518	0.548
Adult								
From 18 to 70 years (45–70 kg)	Drinking water	2000 mL	3.040	3.080	3.640	3.760	6.400	6.800
	Cow milk	350 mL	0.017	0.015	0.023	0.018	0.042	0.040
	Greens	250 g	0.395	0.363	0.575	0.580	1.393	1.920
	Grains	200 g	0.288	0.314	0.518	0.552	0.842	0.822
	Rice	1000 g	0.450	0.480	0.510	0.540	0.690	0.730
Old								
Above 70 years (50–70 kg)	Drinking water	1500 mL	2.280	2.310	2.730	2.820	4.800	5.100
	Cow milk	300 mL	0.014	0.013	0.020	0.015	0.036	0.034
	Greens	200 g	0.316	0.290	0.460	0.464	1.114	1.536
	Grains	150 g	0.216	0.236	0.389	0.414	0.632	0.617
	Rice	1000 g	0.450	0.480	0.510	0.540	0.690	0.730

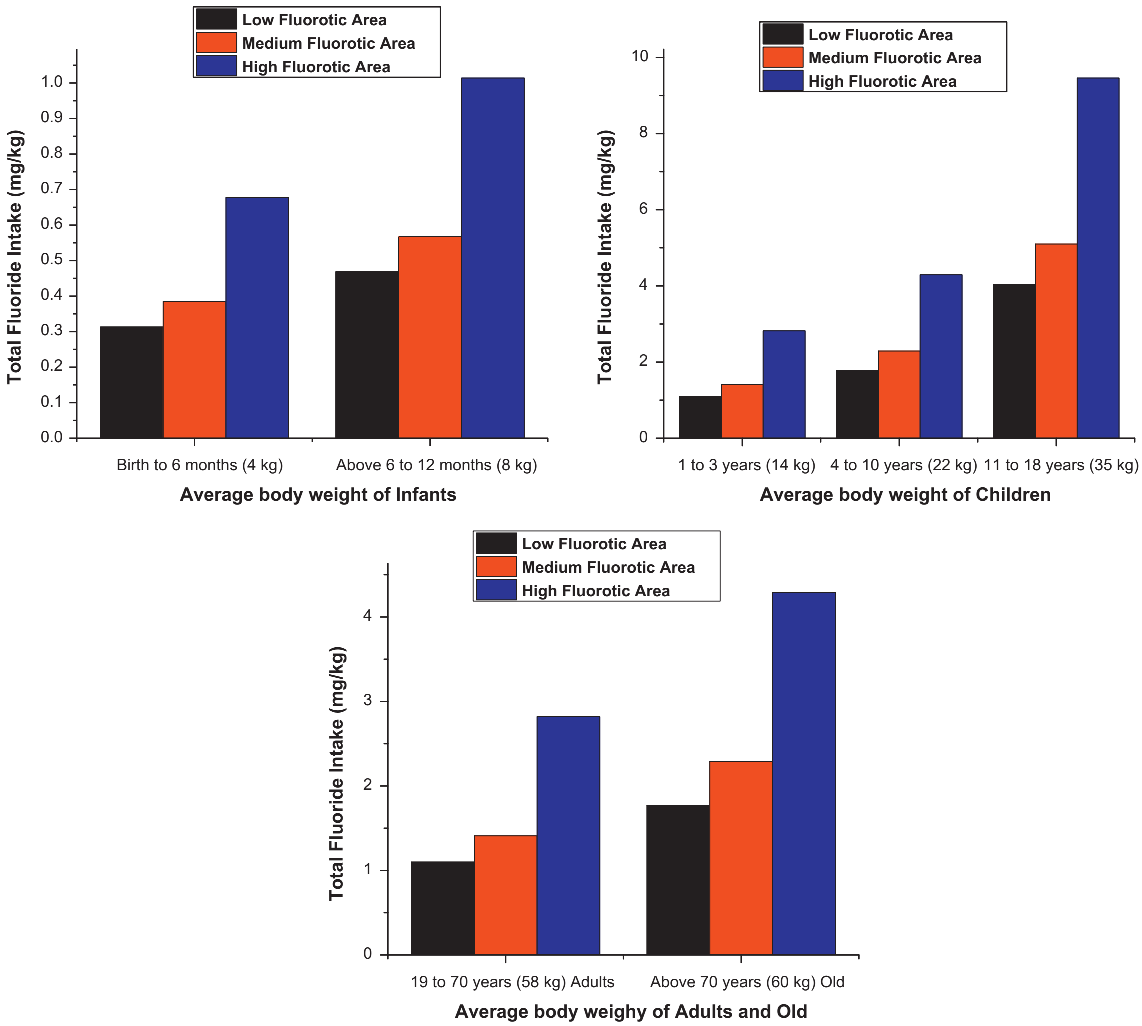

Fig. 2. Average body weight vs total fluoride intake of the respective age group of population.

etc. which are still traditionally grown have high fluoride content [31].

All individual sources of exposure are important since it is the total fluoride intake from all sources that is critical in the development of fluorosis [32]. Among various sources, infant foods, milk formulas, foods containing chicken, some bottled waters and beverages, were identified as significant sources of ingested fluoride by many researchers [33–37]. Fluoride is entering human food chain in increasing amount through the consumption of tea, wheat, spinach, cabbage, carrots and other Indian foods [38–40]. The observations from studies done in China suggested that food can significantly contribute to the total fluoride uptake [41,42].

In Japan, it has been observed that rice and green leafy vegetables (GLVs) main staples in this country produced in polluted areas contained fluoride that is several to more than ten times higher than that found in non-contaminated products [43]. The absorption rate of fluorides contained in rice and GLVs produced in fluoride-polluted areas was greater than 90% approximating that of NaF [44,45]. Sorghum and ragi are consumed extensively in several areas in southern parts of India, where endemic fluorosis is rampant [46]. It is reported that in these areas, the incidence and severity of fluorosis were higher when the staple foods were sorghum or ragi, rather than rice. Rice (*Oryza saliva*) is a major dietary staple of nearly half the world's population. About 95% of this cereal is produced and consumed in South East Asian

Table 11
Correlation between total fluoride intake per day and various dietary sources.

Variables	*r*	R^2	Regression equation	95% confidence interval for *r*	Significant level (*P*)	*F* ratio
Drinking water	0.9991	0.9982	$0.2042 + 0.3264x$	0.9914–0.9999	<0.0001	2155.58
Cow milk	0.9670	0.9351	$-0.0054 + 0.0128x$	0.7222–0.9965	0.0016	57.65
Green leafy leaves	0.9876	0.9753	$-2.5376 + 0.9717x$	0.8868–0.9987	0.0002	157.88
Food grains	0.9536	0.9094	$0.0597 + 0.439x$	0.6282–0.9951	0.0032	40.14
Cooked foods	0.9939	0.9878	$0.2916 + 0.0444x$	0.9429–0.9994	<0.0001	324.13

countries, including India. It is estimated that more than half of the population of India subsists on rice [47]. In rural India, home-made parboiled rice is consumed even in many villages where fluorosis is endemic. The economically backward people, who are the main victims of fluorosis [46], resort to parboiling paddy at the household level. For this purpose, invariably, they use the same source of locally available water which may have unsafe levels of fluoride. It seems possible that such a practice may enhance the fluoride concentration of rice. This study also shows that, the fluoride intake from cooked rice is about 6%–8% in medium and high fluorotic villages, respectively.

Calculated daily intake of fluoride from water, green leafy vegetables, food grains, cow milk and cooked rice in high fluorotic areas of the study shows 3–5 times higher fluoride than 2 mg/d that was estimated as worldwide average for fluoride intake in children and adolescent. For children and adolescents during their developmental age, fluoride daily threshold of 2 mg was recommended by the World Health Organization [3].

Cow milk is a suitable alternative for human milk; fluoride content of samples of cow milk analyzed in this study found to range from 0.04 mg/L to 0.15 mg/L. Fluoride exposure level through milk infants in low fluorotic areas and high fluorotic areas is found to be 2.1% and 2.4%, respectively. Even in high fluorotic area cow milk samples do not have excess fluoride.

The WHO [3] stated that "The most serious effect is the skeletal accumulation of fluoride from long-term excessive exposure to fluoride and its effect on non-neoplastic bone disease specifically, skeletal fluorosis and bone fractures". There is clear evidence from India and China that skeletal fluorosis and an increasing risk of bone fracture occurred at total intake of 14 mg fluoride/d and evidence suggestive of an increasing risk of bone effect at a total intake of about 6 mg fluoride/d [48]. From the results obtained in this study and previous studies, it is evident that after nearly 30 years of experience, the defluoridation system does not provide a safe and dependable supply of domestic water [48]. As a defluoridation plant using activated alumina was installed in one of the villages of the study area in the year 2001 with the financial support from Tamil Nadu water and drainage (TWAD) board, a government institution concerned with provision of safe drinking water is not properly utilized by the people of the area. Another drawback is that the plant is not properly monitored and necessary periodical regeneration of the adsorbent is not carried out.

Adults in high fluorotic areas were exposed to 2 times higher fluoride intake per day than the recommended level of 4 mg/d through all the selected diet sources analyzed. Fluoride levels of all selected diet sources analyzed in medium and high fluorotic areas are nearly 2 and 3 times higher, respectively, than the low fluorotic area diet. The drinking water fluoride level has significant influence on fluorosis prevalence in the study area as it contributes around 66% of the total fluoride intake per day. If water used for drinking or food processing has fluoride level more than 0.65 mg/L, it enhances the daily total fluoride intake more than the standard limit of 4 mg/d for normal adults recommended by the Agency for Toxic Substance and Disease Registry [49,50]. Our findings regarding fluoride levels of food grains and green leafy vegetables show that foods grown in high fluorotic areas have significantly higher fluoride than those grown in low fluorotic area.

5. Conclusion

Planning the supply of low-fluoride water for a fluoride endemic community requires a thorough knowledge of local hydro-geological conditions. An exhaustive inventory of the water sources being used by the community is essential. This study identified water as the major causative factor for fluoride toxicity among other sources, such as cow milk, food grains, green leafy vegetables and cooked rice though, these are consumed in larger quantity by the people, their contribution is less towards the fluoride toxicity of the area studied. Supply of drinking water with less fluoride is emphasized. Rainwater harvesting techniques should be promoted since they have a dilution effect on the fluoride concentration of the ground water of the affected villages.

Acknowledgment

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